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(54) Title: **LOW VISCOSITY HOT-MELT ADHESIVE COMPOSITION FOR NON-WOVENS**

(57) Abstract: Low hot-melt adhesive compositions used in non-woven assemblies, comprising: a) 100 parts by weight of a block copolymer of the formula S-(I/B)-S, wherein S represents a predominantly poly(styrene) block and (I/B) represents a polymer block obtained by at random copolymerization of a mixture of predominantly isoprene and butadiene in a weight ratio in the range of from 60:40 to 40:60, wherein the total block copolymer has an apparent molecular weight in the range of from 124.000 to 145.000 and has a poly(styrene) content of from 30 to 34 wt% and a coupling efficiency of from 60 to 100 %, b) from 250 to 300 parts by weight of a tackifying resin, c) from 5 to 150 parts by weight of a plasticizer, and d) from 1 to 3 parts by weight of auxiliaries, such as stabilizers and/or antioxidants, and disposable articles, comprising at least a non-woven element and assembled by the use of said low hot-melt adhesive compositions as well as block copolymers used therein.

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Description

LOW HOT-MELT VISCOSITY ADHESIVE COMPOSITION FOR NON-WOVENS

Technical Field

- [001] The present invention relates to a low hot-melt viscosity adhesive composition for non-wovens.
- [002] More in particular the present invention relates to a low hot-melt viscosity adhesive composition for the manufacture of sanitary articles such as infant and adult diapers, sanitary napkins, incontinent pads, bed pads, feminine pads, panty shields, and the like, comprising at least one element of non-woven material.

Background Art

- [003] It is generally known in the art, e.g. from WO 9102039 --, EP 0532831 A --, and EP 0802251 A --, that block copolymers, comprising terminal poly(styrene) blocks and one or more central poly(isoprene) blocks and more in particular triblock copolymers, are used in hot melt adhesive compositions for the manufacture of disposable articles. More in particular triblock copolymers having a poly(styrene) content of from 25 to 35 wt% and having a total apparent molecular weight of from 140.000 to 145.000, like KRATON® D-1165 block copolymer, are used in hot-melt adhesive compositions in the diaper industry with two functions:
1. as assembly adhesive to glue the diaper poly(ethylene) main frame and
 2. as an attachment adhesive to glue the elastic attachments which are used as waist and leg bands.
- [004] Although said block copolymers like KRATON® D-1165 block copolymer, show an acceptable hot-melt viscosity/temperature profile, i.e. a sufficient low viscosity at the usually applied hot-melt temperatures of up to 150°C to enable efficient processing, there is still a need for lower viscosities of such hot-melt compositions for economical reasons.
- [005] It will be appreciated that relatively low hot-melt viscosity enables a high production speed in a diaper manufacturing line without significant amounts of products which are out of specification and which have actually to be regarded as waste, which increases the cost price per unit.

- [006] Problems which normally cause said cost price increase may be for example adhesive degradation, damage to the polyethylene film or formation of char.
- [007] Due to the still existing need to improve the economics of the diaper manufacture and to lower the cost price per unit, significant research and development efforts have been and are made to reach these goals.
- [008] Therefore, it is an object of the present invention to provide hot-melt adhesive compositions which show a significantly decreased viscosity at temperatures in the range of from 110 to 150°C in order to avoid the hereinbefore mentioned problems.
- [009] Another object of the present invention is formed by sanitary articles, comprising at least one non-woven layer and manufactured by means of said improved hot-melt adhesive compositions.
- [010] As result of extensive research and experimentation it has now surprisingly been found that linear block copolymers comprising at least two terminal, predominantly poly(styrene) blocks and at least one central (I/B) block, and having corresponding molecular parameters as the until now preferably applied S-I-S block copolymers, show a lower hot-melt viscosity in adhesive compositions at temperatures to be preferably applied in the diaper industry, i.e. in the range of from 110 to 150°C, than the conventional adhesive compositions based on S-I-S block copolymer.
- [011] Moreover, hot melt adhesive compositions comprising said S-(I/B)-S block copolymers have been found to show an increased cohesion.

Disclosure of Invention

- [012] Accordingly the present invention relates to a low hot-melt viscosity adhesive composition used in non-woven assemblies, comprising:
- a) 100 parts by weight of a block copolymer of the formula S-(I/B)-S, wherein S represents a predominantly poly(styrene) block and (I/B) represents a polymer block obtained by at random copolymerization of a mixture of predominantly isoprene and butadiene in a weight ratio in the range of from 60:40 to 40:60 wherein the total block copolymer has an apparent molecular weight in the range of from 124.000 to 145.000 and has a poly(styrene) content of from 30 to 34 wt% and a coupling efficiency of from 60 to 100 %,
 - b) from 250 to 300 parts by weight of a tackifying resin,
 - c) from 5 to 150 parts by weight of a plasticizer , and
 - d) from 1 to 3 parts by weight of auxiliaries, such as stabilizers and/or antioxidants.
- [013] Said adhesive compositions comprising for instance 25 wt% of block copolymer in its formulation have appeared to show a hot-melt viscosity at about 130°C, which is at least 25% lower (e.g. 20 Pas) than a corresponding adhesive composition having the same composition, but comprising a corresponding block copolymer of the S-I-S type.

[014] It will be appreciated that a significant advantage therefore has been reached, as the compositions of the present invention enable either processing at lower temperatures (i.e. in the range of from 110 to 130°C) or faster processing.

[015] Other aspects of the present invention are formed by disposable articles such as infant and adult diapers, sanitary napkins, incontinent pads, bed pads, feminine pads, panty shields, comprising at least one non-woven element and assembled by the use of the hereinbefore specified low hot melt viscosity adhesive compositions, and by the specific block copolymers to be used therein.

[016] A particular aspect of the present invention is formed by feminine pads, using the hereinbefore specified low hot melt viscosity compositions as positioning adhesive.

Mode for the Invention

[017] The main block copolymer used in the adhesive composition is

[018] component (a)

[019] A block copolymer, having a structure represented by the general formula S-(I/B)-S, optionally mixed with a diblock copolymer S-(I/B), wherein S represents a predominantly poly(styrene) block, (I/B) represents a block of a randomly copolymerized mixture of predominantly isoprene and butadiene, wherein the weight ratio between isoprene and butadiene is in the range of from 60:40 to 40:60 (or in a molar ratio of from 0.89/0.75 to 0.75/0.89). More preferred weight ratios of isoprene and butadiene are in the range of from 55:45 to 45:55.

[020] With the terms "predominantly poly(styrene)" and "mixtures of predominantly isoprene and butadiene" are meant that in addition to the main monomer, i.e. styrene on the one hand and on the other hand isoprene and butadiene, one or more other minor comonomers may be present in the starting monomer in small amounts, i.e. up to 5 wt% of copolymerizable monomer. However, preferably substantially pure (co)monomers may be used for the preparation of the respective blocks.

[021] Examples of minor comonomers of used in addition to styrene are alpha-methylstyrene, p-methylstyrene, o-methylstyrene, p-tert.butylstyrene, dimethylstyrene and vinyl naphthalene or mixtures thereof.

[022] The mixed polymer midblock (I/B) is made of butadiene and isoprene as copolymerizing monomers, although it too may contain minor amounts of other comonomers, e.g. up to 5 wt% of a copolymerizable monomer such as styrene (based on the weight of the total block), but mixtures of substantially pure isoprene and butadiene are preferred. Additional examples of comonomers used in addition to the isoprene/butadiene mixture include the aforementioned comonomers of styrene as well as neoprene or chloroprene.

[023] In the block copolymers according to the present invention, the poly(styrene)

content (which may include optional comonomers) is in the range of 25-40 wt%, preferably 30-34 wt%, based on the total block copolymer. The proportion of bound butadiene is 18-80 wt%, preferably 40-70 wt% in total. The proportion of bound isoprene is 15-70 wt%, preferably 30-70 wt%. These amounts of bound monomers (plus copolymerizable monomers, if any) add up to 100 wt%.

[024] These polymer blocks S preferably have a true molecular weight in the range from 9,500 to 25,000 and more preferably from 14,000 to 15,000.

[025] The block copolymers to be applied in the adhesive compositions according to the present invention each preferably have a weight average molecular weight (M_w , expressed in terms of polystyrene) ranging from 124,000 to 145,000, more preferably from 124,000 to 145,000 as determined by Gel Permeation Chromatography (GPC, analogous to the method described in ASTM D5296-97).

[026] The block copolymers to be applied in the adhesive compositions according to the present invention each preferably contain 1,2-vinyl bonds and/or 3,4-vinyl bonds in a proportion in the range of from 4 to 10 wt%, based on the weight of the conjugated diene (or in the range of from 0.08 to 0.70 mole/mole %).

[027] Preferably the (I/B) block has a vinyl content in the polymerized butadiene in the range of from 7 to 9 wt% and a vinyl content in the polymerized isoprene in the range of from 4 to 6 wt%.

[028] Said block copolymers to be applied as main component (a) in the adhesive composition, have a randomly copolymerized block (I/B), which means that the mixed midblock shows no significant single homopolymer block formation.

[029] They can be prepared as described in WO 02057386 --.

[030] More in particular, polymers having midblocks may be defined as having average homopolymer block lengths of less than 100 monomer units, preferably less than 50 monomer units, more preferably less than 20 monomer units.

[031] Average homopolymer block length may be determined by carbon-13 NMR, as described in detail in WO 02057386 --.

, which is herein incorporated by reference.

[032] Although the hereinbefore mentioned publication WO 02057386 --.

disclosed adhesive compositions comprising one or more styrenic block copolymers comprising a central (I/B) block, a tackifier resin and one or more plasticizers, there was not any teaching to a person skilled in the art as to the presently found decrease in viscosity of hot-melt adhesive compositions in comparison to the conventional S-I-S

block copolymer containing hot-melt adhesive compositions.

[033] In this respect particular reference is made to the block copolymer F in WO 02057386 page 19, Table 2, which is actually the same as comparative block copolymer B in the tables 2-4 in the present application and which did not provide the presently claimed improved low hot-melt viscosity in adhesive compositions for non-wovens. Therefore a person skilled in the art could in no way expect these improved properties of the specifically selected block copolymers in combination with specific weight ratios of tackifying resin plasticizers and auxiliaries.

[034] Moreover, from DE 2942128 --.

a hot-melt adhesive composition was known, comprising (a). 100 parts by weight of a non-hydrogenated linear block copolymer with a molecular weight of 30,000-300,000 of the formula A-B-A. wherein the terminal A blocks of poly(styrene) represent from 10 to 50 wt% of the block copolymer, wherein the block B is an elastomeric statistical copolymer of butadiene and isoprene in a weight ratio of from 70:30 to 5:95. However said applied block copolymers did actually not contain substantial proportions of diblock copolymer.

No suggestion was provided to reach the further improved low hot-melt viscosity adhesive composition for non-wovens by selecting block copolymers having specific molecular weights and coupling efficiencies or diblock contents. In this respect reference is particularly made to the absence of significant amounts of diblock copolymer in the actually prepared block copolymers A-B-A.

[035] From JP 5345885 --.

adhesive compositions were known which comprised a styrene-isoprene/butadiene-styrene block copolymer having an isoprene content in the isoprene/butadiene block of from 10 to 90 wt%, a poly(styrene) content in the range of from 5 to 50 wt% and a molecular weight of the poly(styrene) blocks in the range of from 5,000 to 30,000. However, not any teaching can be found in said document enabling to reach low viscosity hot-melt stable adhesive compositions presently aimed at. More in particular no suggestion was provided to reach the further improved low hot-melt viscosity in adhesive compositions for non-wovens as presently claimed by selecting specific values for the molecular weight and the coupling efficiency or diblock content in the block copolymers to be applied.

[036] The block copolymers according to the present invention can be made e.g. by coupling living diblock copolymer prepared by anionic polymerization with a coupling agent.

[037] As examples of the coupling agent, may be mentioned tin coupling agents;

halogenated silicon coupling agents; alkoxysilanes; divinyl aromatic compounds; halogenated alkanes; halogenated aromatic compounds; epoxy compounds such as the diglycidyl ether of bisphenol-A and the like and other coupling agents such as benzoic esters, CO_2 , and 1-chloro-1,3-butadiene.

[038] The main block copolymer in component (a) may hence comprise a mixture of the coupled polymer according to the general formulae S-(I/B)-S and of the intermediate diblock S-(I/B), e.g. in a weight ratio of 100/0 to 60/40, and preferably from 100/0 to 70/30.

[039] It will be appreciated that the main block copolymer component (a) may also be formed by a block copolymer obtained by sequential polymerization of batches of the respective monomers (e.g. styrene and mixtures of butadiene/isoprene, optionally in combination with reinitiation, if additional diblock copolymer is desired).

[040] The main block copolymer component (a) can be made by mere adaptation of common processes used for the preparation of S-B-S type block copolymers and/or S-I-S type block copolymers, using a mixture of butadiene/isoprene instead. Of importance in the preparation of the block copolymers to be used according to the present invention is to avoid homopolymer-block formation to ensure the desired (I/B) ratio.

[041] It will be appreciated that another aspect of the present invention is formed by the block copolymers specified herein before and in the claims.

[042] Component (b)

[043] The tackifying resins to be used in the hot-melt adhesive compositions of the present invention can be selected from a great variety of resins known to be applicable in hot-melt adhesives.

[044] They can be selected from modified aliphatic hydrocarbon resins such as modified C_5 hydrocarbon resins (C_5/C_9 resins), styrenated terpene resins, fully or partially hydrogenated C_9 hydrocarbon resins, hydrogenated cycloaliphatic hydrocarbon resins, hydrogenated aromatic modified cycloaliphatic hydrocarbon resins, and mixtures thereof.

[045] More preferred examples of resins to be used as component (b) are: water-white hydrocarbon resins of the ESCOREZ™ resin series, such as ESCOREZ 5600, ESCOREZ 5400, ESCOREZ 5300 and the like, or hydrocarbon resins of the REGALITE™ resin series such as REGALITE-1090, REGALITE-7100 and REGALITE S-5100 resins and the like, or the ARKON™ resin series, like ARKON P and M resin.

[046] Preferred solid tackifying resins will have Ring and Ball softening points in the range of from 90°C to 105°C and will have an aromatic H-NMR content between 0 and 16 % and more preferably between 0 and 12 %.

[047] In preferred low hot-melt viscosity adhesive compositions of the present invention the component (b) is applied in amounts of from 250 to 280 parts by weight of

tackifying resin per 100 parts by weight of block copolymer components (a) and (b).

[048] Component (c)

[049] Suitable plasticizers include predominantly plasticizing oils that are paraffinic or naphthenic in character (carbon aromatic distribution $\leq 5\%$, preferably $\leq 2\%$, more preferably 0% as determined according to DIN 51378) and a glass transition temperature lower than -55°C as measured by Differential Scanning Calorimetry. Those products are commercially available from the Royal Dutch Shell Group of companies, like SHELLFLEXTM, CATENEXTM, and ONDINATM oils. Other oils include KAYDOLTM oil from Witco, or TUFFLOTM oils from Arco or NYPLASTTM from NYNAS. Other plasticizers include compatible liquid tackifying resins like REGALREZTM R-1018 or WINGTACK 10.

[050] Other plasticizers may also be added, like olefin oligomers; low molecular weight polymers ($\leq 30,000$ g/mol) like liquid polybutene, liquid polyisoprene copolymers, liquid styrene/isoprene copolymers or liquid hydrogenated styrene/conjugated diene copolymers; vegetable oils and their derivatives; or paraffin and microcrystalline waxes.

[051] The hot-melt adhesive composition according to the present invention comprises a plasticizer in a weight proportion of from 11 to 27 wt%, relative to the weight of the complete composition and of from 5 to 150 parts by weight of plasticizer per 100 parts by weight of block copolymer constituent (a). Preferably the hot-melt adhesive composition comprises from 100 to 125 parts by weight of plasticizer per 100 parts by weight of block copolymer.

[052] It will be appreciated that each block copolymer of component (a) may be pre-blended with a small amount of plasticizer by the manufacturer of said copolymer.

[053] Other components (non-limitative)

[054] Other rubber components may be incorporated into the adhesive compositions according to the present invention. It is also known in the art that various other components can be added to modify the tack, the odour, the colour of the adhesives.

[055] Antioxidants and other stabilizing ingredients can also be added to protect the adhesive from degradation induced by heat, light and processing or during storage. Several types of antioxidants can be used, either primary antioxidants like hindered phenols or secondary antioxidants like phosphite derivatives or blends thereof.

[056] Examples of commercially available antioxidants are IRGANOXTM 565 (2,4-bis-(n-octylthio)-6-(4-hydroxy-3,5-di-tertiary-butyl anilino)-1,3,5-triazine) and IRGANOX 1010 from Ciba-Geigy (tetrakis-ethylene-(3,5-di-tertiary-butyl-4-hydroxy-hydrocinnamate)methane) and POLYGARDTM HR from Uniroyal (tris-(2,4-di-tertiary-butyl-phenyl)phosphite). Other antioxidants developed to protect the gelling of the polybutadiene segments can also be

use, like the SUMILIZER™ GS (2[1-(2-hydroxy-3,5-di-ter-pentylphenyl)ethyl] - 4,6-di-ter-pentylphenylacrylate) and SUMILIZER T-PD from Sumitomo (pentaerythrythyltetrakis(3-dodecylthiopropionate)); or mixtures thereof.

[057] Preparation of the composition

[058] No particular limitation is imposed on the preparation process of the adhesive composition. Therefore, there may be used any process such as a mechanically mixing process making use of rolls, a Banbury mixer or a Dalton kneader, a hot-melt process characterized in that heating and mixing are conducted by using a melting kettle equipped with a stirrer, like a turbo-mixer, a high shear Z-blade mixer or a single- or twin-screw extruder, or a solvent process in which the compounding components are poured in a suitable solvent and stirred, thereby obtaining an intimate solution of the pressure sensitive adhesive composition.

[059] Use of the composition

[060] Hot-melt adhesive compositions according to the present invention are efficiently applied as assembly adhesive composition or as attachment adhesive composition for assembling sanitary articles and in particular diapers, comprising a polyethylene framework and at least one non-woven lining element.

[061] The present invention will hereinafter be illustrated more specifically by the following examples, however without restricting its scope to these specific embodiments.

[062] Test methods

- **Polystyrene content** was determined by ¹H-NMR.
- **Average homopolymer block lengths** were determined by ¹³C NMR, using the method described hereinbefore. ¹³C NMR spectra were obtained by using a BRUKER AMX-500 FT spectrometer operating at 125 MHz. Quantitative proton-decoupled spectra were recorded with a 90° ¹³C excitation pulse and a repetition rate of 10 s. Polymer solutions in CDCl₃, having a concentration of 10 % (w/w) were used. To improve the relaxation time 0.1 mol/l chromium acetyl acetonate was added. The applied broadening was 2 Hz. The spectra were referred such that the aliphatic carbons of trans-polybutadiene were 31.9 ppm.
- **Hot-melt viscosity** is measured with a rotational viscometer following ASTM D-3236-78. The viscosity is measured by the torque required to rotate a spindle at constant speed while immersed in a fluid. A sample is placed in a RVTDV-II equipped with a Brookfield Thermocell and the hot-melt viscosity is measured in a temperature range of 110°C to 160°C. Results are expressed in Pascal.second (Pa.s).

[063] Synthesis of the block copolymers A to H

[064] Cyclohexane, styrene, butadiene and isoprene were purified by activated aluminumoxide and stored at 4°C under a nitrogen atmosphere. EPON™ 826 diglycidyl ether was used as coupling agent. Prior to synthesis, a monomer mixture of butadiene and isoprene (at a weight/weight ratio given in Table 1) was prepared and stored under nitrogen at 4°C. This mixture was used as such.

[065] An autoclave, equipped with a helical stirrer was charged with cyclohexane and the content was heated to 50 to 60°C. As initiator sec-BuLi was dosed immediately followed by styrene monomer, which was allowed to polymerize to completion. The reaction temperature was increased to 70°C, at which temperature a butadiene/isoprene monomer mixture (B/I) was dosed and reacted. The resulting diblock was coupled with an excess EPON 826 diglycidyl ether or 1,2-dibromoethane (EDB). This excess was optionally scavenged with sec-BuLi and followed by addition of ethanol as terminator. The reaction mixture was cooled to 40°C, transported to a blending vessel and a stabilization package was added (comprising IRGANOX 565 and tris(nonylphenol)phosphite 0.08/0.35 phr as a cyclohexane solution) and stirred at RT. Dry rubber was obtained by steam coagulation finishing, followed by drying in an oven.

[066] Table 1 lists the amounts in which the components A and B have been used. The polymers were analyzed by GPC and the results thereof are in Table 2.

[067] Polymers A, C, D, E, F, G, and H have been coupled with EPON 826. Polymer B, included as comparative with a MW greater than 150,000, has been coupled with EDB.

[068] These polymers can also be polymerized with the hereinbefore described sequential/reinitiation technology.

[069]

Table 1

Polymer	A	B
Cyclohexane (l)	77	6
Initiator (mmol)	20.5	17.5
Styrene (gram)	638	256.7
B/I (ratio)	1	1
B/I (gram)	1490	598.1
EPON 826 (gram)	0.24	-
EDB (mol%)	-	21
Ethanol (ml)	1	1.2

[070]

Table 2

Polymer	A	B°	C	D	E	F	G	H
Mw Polystyrene*10 ³	14.2	14.8	14.4	13.7	14.2	13.9	14.2	14.6
Total Mw*10 ³	143	153	137	138	142	124	128	130
Coupling efficiency %	81	81	80	79	73	73	81	79
Polystyrene content wt%	31	30	30	30	31	34	34	33
Bd/Ip ratio wt/wt	1	1	1	1.45	0.98	0.69	0.72	0.99
vinyl in Bd wt %	8	8	8	8	8	8	8	8
vinyl in Ip wt%	5	5	5	5	5	5	5	5

[071] ° comparative block copolymer, not according to the invention.

Further components used in the tested adhesive compositions have been listed in Table 3.

[072]

Table 3

<p>Kraton D-1164 is a linear styrene-isoprene-styrene block copolymer with about 30 wt% polystyrene content, a total molecular weight of about 126,000 g/mole and a coupling efficiency of 100%</p> <p>Kraton D-1165 is a linear styrene-isoprene-styrene block copolymer with 30 wt% of polystyrene content, a coupling efficiency in the range of 78-84% and a total molecular weight in the range of 130,000-152,000 g/mole.</p> <p>Polymer A is a linear styrene-isoprene/butadiene-styrene block copolymer with 31 wt% polystyrene content, a coupling efficiency of 81 %, a total molecular weight of 143,000 g/mole and a isoprene/butadiene w% ratio of 50/50.</p> <p>Polymer B is a linear styrene-isoprene/butadiene-styrene block copolymer with a polystyrene content of 30 wt%, a coupling efficiency of 86 %, a total molecular weight of 154,000 g/mole and an isoprene/butadiene wt% ratio of 50/50. Polymer B actually is to be considered as comparative block copolymer.</p> <p>Polymer C is a linear styrene-isoprene/butadiene-styrene block copolymer with 30 wt% of polystyrene content, a coupling efficiency of 79.9 %, a total molecular weight of 137,000 g/mole and an isoprene/butadiene wt% ratio of 50/50.</p> <p>Polymer D is a linear styrene-isoprene/butadiene-styrene block copolymer with 30.3</p>

wt% of polystyrene content, a coupling efficiency of 79.4 %, a total molecular weight of 138,000 g/mole and an isoprene/butadiene wt% ratio of 40/60

Polymer E is a linear styrene-isoprene/butadiene-styrene block copolymer with 30.6 wt% of polystyrene content, a coupling efficiency of 73 %, a total molecular weight of 141,700 g/mole and an isoprene/butadiene wt% ratio of 50.5/49.5

Polymer F is a linear styrene-isoprene/butadiene-styrene block copolymer with 33.6 wt% of polystyrene content, a coupling efficiency of 72.8 %, a total molecular weight of 124,200 g/mole and an isoprene/butadiene wt% ratio of 59/41

Polymer G is a linear styrene-isoprene/butadiene-styrene block copolymer with 33.9 wt% of polystyrene content, a coupling efficiency of 80.7 %, a total molecular weight of 127,900 g/mole and an isoprene/butadiene wt% ratio of 58/42.

Polymer H is a linear styrene-isoprene/butadiene-styrene block copolymer with 33 wt% of polystyrene content, a coupling efficiency of 79.3 %, a total molecular weight of 129,500 g/mole and an isoprene/butadiene wt% ratio of 50/50

REGALITE-7100 is a hydrocarbon resin from Eastman

REGALITE-1090 is an hydrocarbon resin from Eastman

ONDINA 68 is a paraffinic oil from Shell

IRGANOX 1010 is an antioxidant from CIBA

[073] Example 1

[074] The viscosity of hot-melt adhesive compositions comprising the ingredients as specified hereafter was measured at different temperatures.

[075] Each composition comprised:

100 parts by weight of block copolymer,
250 parts by weight of REGALITE-1090 hydrocarbon resin,
50 parts by weight of ONDINA 68 plasticizing oil,
3 parts by weight of antioxidant (IRGANOX 1010).

[076]

Table 4 (Viscosities in Pa.s)

Temp.	D-1165	A	B°	C	D	E	F	G	H
130°C	79.4	25.7	110	25	33.4	31	16.3	29.4	23.4
140°C	20.7	10.2	26.6	11.1	14.7	12.8	6.9	11.4	10.7
150°C	7.6	5.5	12	6.8	8.3	7.2	3.8	6	6.1
160°C	3.6	3.6	8.2	4.4	5.3	4.6	2.4	3.6	4

[077] ° comparative block copolymer outside the present invention.

[078] Example 2

[079] The viscosity of hot-melt adhesive compositions comprising the ingredients as specified hereafter was measured at different temperatures.

[080] Each composition comprised:

100 parts by weight of block copolymer,

272 parts by weight of hydrocarbon resin REGALITE-7100,

125 parts by weight of ONDINA 68 plasticizing oil,

3 parts by weight of antioxidant IRGANOX 1010

[081]

Table 5 (Viscosities in Pa.s)

Temperature	D-1164	D-1165	A
110°C	26.6	42.3	20.4
120°C	11.32	13.7	9.45
130°C	5.51	6.27	5.0
140°C	3.22	3.38	3
150°C	2.02	2.07	2
160°C	1.33	1.38	1.38

Claims

- [001] Low hot-melt adhesive composition used in non-woven assemblies, comprising
- a) 100 parts by weight of a block copolymer of the formula S-(I/B)-S, wherein S represents a predominantly poly(styrene) block and (I/B) represents a polymer block obtained by at random copolymerization of a mixture of predominantly isoprene and butadiene in a weight ratio in the range of from 60:40 to 40:60, wherein the total block copolymer has an apparent molecular weight in the range of from 124.000 to 145.000 and has a poly(styrene) content of from 30 to 34 wt% and a coupling efficiency of from 60 to 100 %,
 - b) from 250 to 300 parts by weight of a tackifying resin,
 - c) from 5 to 150 parts by weight of a plasticizer , and
 - d) from 1 to 3 parts by weight of auxiliaries, such as stabilizers and/or antioxidants.
- [002] Low hot-melt viscosity adhesive composition according to claim 1, wherein the (I/B) block has a vinyl content in the polymerized butadiene in the range of from 7 to 9 wt% and a vinyl content in the polymerized isoprene in the range of from 4 to 6 wt%.
- [003] Low hot-melt viscosity adhesive composition according to claim 1, wherein from 250 to 280 parts by weight of hydrocarbon resin per 100 parts by weight of block copolymer have been included.
- [004] Disposable articles such as infant and adult diapers or sanitary napkins, incontinent pads, bed pads, feminine pads, panty shields, comprising at least one non-woven element and assembled by the use of the low hot-melt viscosity adhesive composition according to claims 1-3.
- [005] Block copolymers of the formula S-(I/B)-S, wherein S represents a predominantly poly(styrene) block and (I/B) represents a polymer block obtained by at random copolymerization of a mixture of predominantly isoprene and butadiene in a weight ratio in the range of from 60:40 to 40:60, wherein the total block copolymer has an apparent molecular weight in the range of from 124.000 to 145.000 and has a poly(styrene) content of from 30 to 34 wt% and a coupling efficiency of from 60 to 100 %.
- [006] Block copolymers according to claim 5, wherein the S blocks have been derived from pure styrene.
- [007] Block copolymers according to claim 6, wherein the (I/B) block has a vinyl content in the polymerized butadiene in the range of from 7 to 9 wt% and a vinyl content in the polymerized isoprene in the range of from 4 to 6 wt%.

INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP2005/050614

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C09J153/02 C08F297/04 A61L15/58

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C09J C08L C08F A61L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 02/057386 A (DE KEYZER NOEL RAYMOND MAURICE ; VAN DIJK MARTIN (NL); KRATON POLYMERS) 25 July 2002 (2002-07-25) cited in the application claims 1,4-6,9 table 2 page 1, line 6 - line 14	1-7
X	DE 29 42 128 A (BASF AG) 30 April 1981 (1981-04-30) claim 1	1-7
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Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

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Date of the actual completion of the international search

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INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP2005/050614

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	<p> DATABASE WPI Section Ch, Week 199405 Derwent Publications Ltd., London, GB; Class A18, AN 1994-040113 XP002216588 & JP 05 345885 A (KURARAY CO LTD) 27 December 1993 (1993-12-27) abstract </p>	1-7

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP2005/050614

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
WO 02057386	A	25-07-2002	BR 0206601 A	10-02-2004
			CN 1516729 A	28-07-2004
			WO 02057386 A2	25-07-2002
			EP 1354016 A2	22-10-2003
			JP 2004520465 T	08-07-2004
			US 2004116582 A1	17-06-2004
DE 2942128	A	30-04-1981	DE 2942128 A1	30-04-1981
JP 5345885	A	27-12-1993	NONE	

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